

# THERMOMECHANICAL MODEL OF HYDRATION SWELLING IN SMECTITIC CLAYS: II THREE-SCALE INTER-PHASE MASS TRANSFER: HOMOGENIZATION AND COMPUTATIONAL VALIDATION

MÁRCIO A. MURAD<sup>\*,†</sup>

*Rua Getulio Vargas 333 Quitandinha, CP 95113 Petropolis, RJ Brazil*

## SUMMARY

In Part I<sup>1</sup> a two-scale thermomechanical theory of expansive compacted clays composed of adsorbed water and clay platelets was derived using a mixture-theoretic approach and the Coleman and Noll method of exploitation of the entropy inequality. This approach led to a two-scale model which describes the interaction between thermal and hydration effects between the adsorbed water and clay minerals. The purpose of this paper is twofold. Firstly, partial results toward a three-scale model are derived by homogenizing the two-scale model for the clay particles (clusters of clay platelets and adsorbed water) with the bulk water (water next to the swelling particles). The three-scale model is of dual porosity type wherein the clay particles act as sources/sinks of water to the macroscale bulk phase flow. One of the notable consequences of the homogenization procedure is the natural derivation of a generalized inter-phase mass transfer equation between adsorbed and bulk water. Further, variational principles and finite element approximations based on the Galerkin method are proposed to discretize the two-scale model. Numerical simulations of a bentonitic clay used for engineered barrier of nuclear waste repository are performed and numerical results are presented showing the influence of physico-chemical effects on the performance of the clay buffer. Copyright © 1999 John Wiley & Sons, Ltd.

Key words: swelling clay buffer; homogenization; dual porosity; mass transfer; physico-chemical effects; finite element method

## 1. INTRODUCTION

In a earlier paper<sup>1</sup> Hybrid Mixture Theory (HMT) was used to model flow, deformation and heat transfer in two-scale swelling systems such as highly compacted smectitic clays, composed of clay minerals and adsorbed (vicinal) water. By adopting a proper theory of constitution including appropriate internal variables to capture the swelling character of these systems, HMT provided a natural framework for a proper thermodynamic definition of intra-particle thermo-physico-chemical stresses. In particular, HMT provided a rational basis for a thermodynamic derivation

\* Correspondence to: M. A. Murad, LNCC/CNPq, Rua Getulio Vargas 333, Quitandinha, CP 95113, Petropolis, RJ, Brazil

<sup>†</sup> Laboratório Nacional de Computação Científica, LNCC/CNPq, Rua Lauro Muller 455, 22290 – Rio de Janeiro, Brazil

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of some modified Terzaghi's effective stress principles which incorporate physico-chemical effects (e.g. References 2 and 3) within the clay particles. In addition, HMT led to a modified form of Darcy's law for the adsorbed water flow which involves a volume fraction gradient as an additional driving force for vicinal water flow. This component accounts for the hydrophilicity between adsorbed water and the clay minerals and is consistent with the microscopic results of thin liquid films showing that thin films tend to flow following the gradient of their thickness (or disjoining pressure gradient (see e.g. Reference 4). Other consequences of the HMT approach include the appearance of a retardation viscosity coefficient which quantifies the viscoelastic behaviour (relaxation) of the vicinal water and an additional physico-chemical coefficient of thermal expansion which among other effects incorporates the mechanical work of hydration stresses in the energy balance.

The two-scale HMT model of Murad<sup>1</sup> applies to a two-phase swelling system (adsorbed liquid and clay platelets) with a monomodal distribution, in which all the water is adsorbed to the clay surface (e.g. highly compacted clay). The extension of this approach to three-scale swelling systems, i.e. porous swelling clay particles in contact with bulk water requires an additional upscaling and leads to a three-scale model. A three-scale picture (micro, meso and macro) of a swelling clay consisting of porous swelling particles is depicted in Figure 2 of Reference 1. The particles are in contact with one another and bulk water. Each particle consists of clay colloids and adsorbed water. At the microscale the model has two phases, the disjoint clay minerals and the vicinal water. At the mesoscale (the homogenized microscale) the model consists of clay particles (where the two-scale HMT model of Murad<sup>1</sup> holds) and bulk water. The macroscale consists of the bulk water homogenized with the mesoscale particles. In this paper we adopt the homogenization procedure<sup>5</sup> to upscale the two-scale mesoscopic model and consequently derive some three-scale results. Within this upscaling procedure we consider the two-scale HMT thermomechanical equations of Murad<sup>1</sup> governing hydration swelling in the clay particle domain. The complete mesoscopic description is established by coupling the HMT system with the bulk water governing equations (in the bulk phase domain) through boundary conditions on the particle-bulk phase interface. Hence, information is propagated to the macroscale using the homogenization tools. We show that one of the notable consequences of the homogenization procedure is the rigorous derivation of a generalized inter-phase mass transfer function between adsorbed and bulk water. This mass transfer was first postulated by Ma and Hueckel<sup>6-8</sup> as a phenomenological concept of interconstituent mass exchange appearing as a source term in the fluid mass balances. Thus, the proposed approach consists of a first attempt to overcome the difficulties inherent to the necessity of postulating *ad hoc* terms in the governing equations.

We also discuss the numerical approximation of the proposed multiscale model. We concentrate our analysis on the discretization of the two-scale model of Murad<sup>1</sup> by finite elements. We show that the linearized version of this system has a structure similar to the thermoelastic consolidation model of granular (non-swelling) media and exhibits additional physico-chemical and viscous-type terms accounting for the thermo-physico-chemical interaction between the absorbed fluid and the clay minerals. The variational formulations and finite element approximations proposed to the classical thermoelastic consolidation governing equations e.g. Reference 9 are extended to solve the two-scale HMT model. We then perform numerical simulations of a problem related to a clay-based engineered barrier for encapsulating a heat generating container involving highly compacted bentonitic clays. The influence of physico-chemical effects on the performance of the clay buffer is then illustrated numerically.

## 2. TWO-SCALE LINEARIZED MODEL FOR COMPACTED SWELLING CLAY

Consider the clay particles as a mixture of two phases (the solid clay platelets and adsorbed water) viewed as coexisting continua, which undergo independent motions  $\mathbf{x} = \mathbf{x}_\alpha(\mathbf{X}_\alpha, t)$ ,  $\alpha = l, s$  with respect to each reference configuration (here  $\mathbf{x}$  denotes the spatial position of the particle of the  $\alpha$ -phase at time  $t$  with respect to a reference position  $\mathbf{X}_\alpha$ ). Let the subscript  $\alpha = l, s$  denote the adsorbed liquid and solid phase, respectively, and let  $\rho_\alpha$ ,  $\phi_\alpha$ ,  $T_\alpha$  and  $A_\alpha$  denote, respectively, the averaged density, volume fractions, temperature and intensive Helmholtz free energy of the  $\alpha$ -phase ( $\alpha = l, s$ ). Further let the average mesoscopic linearized strain tensor of the solid phase,  $\mathbf{E}_s$ , be given as

$$\mathbf{E}_s = \nabla^s \mathbf{u}_s, \quad (1)$$

where  $\nabla^s \mathbf{u}_s = \frac{1}{2}(\nabla \mathbf{u}_s + \nabla \mathbf{u}_s^T)$ , with  $\mathbf{u}_s$  denoting the displacement of the solid phase. Adopting the same assumptions of Murad<sup>1</sup> consider particles initially at a homogeneous, equilibrium and isotropic state and assume small deformations and small deviations in temperature and density from their reference values. Assume that interfaces contain no thermodynamic properties, entropy fluxes are solely due to heat fluxes, solid and fluid are non-polar, non-reacting phases at local thermal equilibrium ( $T_l = T_s = T$ ), the adsorbed water at the mesoscale is non-viscous, the clay platelets are incompressible, physico-chemical effects are dominated by the hydration forces and the dissipation term in the energy equation and convective effects are neglected. In addition, following Murad,<sup>1</sup> assume that all external sources except the one appearing in the energy equation (heat supply) are absent, and consider the range of moderate moisture contents, wherein the clay minerals are separated by a distance equivalent to 10 to 30 layers of mobile vicinal water. This latter assumption implies that, at equilibrium, clay particles are able to withstand the normal hydrostatic swelling pressure but, on the other hand, deviatoric shear stresses are only supported by effective stresses see Reference 1. By postulating constitutive dependence of the free energies in the form  $A_s = A_s(T, \rho_s, \mathbf{E}_s)$  and  $A_l = A_l(T, \rho_l, \phi_l)$ , using the Coleman and Noll method of exploiting the entropy inequality<sup>10</sup> and linearizing about the initial homogeneous equilibrium state, Murad<sup>1</sup> derived a two-scale thermomechanical model governing the swelling of the clay particles. Let  $\Omega_l$  denote the clay particle domain and let the constant properties at the initial equilibrium state be denoted by the overline superscript (e.g.  $\bar{\rho}_l$  for the adsorbed water density). The two-scale governing equations of Murad<sup>1</sup> valid in  $\Omega_l$  are

*Mass of the solid phase:*

$$\frac{\partial \phi_l}{\partial t} - \bar{\phi}_s \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t} = 0 \quad (2)$$

*Total mass:*

$$\bar{\phi}_l \frac{\partial \rho_l}{\partial t} + \bar{\rho}_l \left( \operatorname{div} \mathbf{q}_l + \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t} \right) = 0 \quad (3)$$

where  $\mathbf{q}_l$  is the mass-average Darcy's velocity of the adsorbed fluid.

*Total momentum:*

$$\operatorname{div} \mathbf{t} = 0$$

where  $\mathbf{t}$  denotes the total stress tensor of the clay particles.

*Pore pressure definition:*

$$p \equiv p_1 \phi_1 + \phi_s p_s, \quad p_1 \equiv \rho_1^2 \frac{\partial A_1}{\partial \rho_1} \quad (4)$$

where  $p$  is the pore pressure (total pressure of the clay particle) and  $p_1, p_s$  the thermodynamic pressures of the adsorbed water and solid phase.

*Near equilibrium viscoelastic relaxation:*

$$p_1 - p_s = \bar{\mu}_* \frac{\partial \phi_1}{\partial t} \quad (5)$$

This shows that near equilibrium, the thermodynamic pressure of the adsorbed fluid and solid phases are not equal. The commonly assumed equality  $p_1 = p_s = p$  for granular non-swelling media may not necessarily hold, especially for swelling systems. The coefficient  $\bar{\mu}_*$  may be thought of as a relaxation factor (or viscosity coefficient) which accounts for the viscoelastic behavior of thin films at the microscale, i.e. the adsorbed fluid molecules tend to redistribute over the interlamellar spaces as they are disturbed from their equilibrium state, consequently leading to a relaxation phenomenon<sup>11</sup>. In reference 12,  $\bar{\mu}_*$  was identified with an excess in the vicinal water viscosity relative to bulk water.

*Modified Terzaghi's effective stress principle:*

$$\mathbf{t} = -p\mathbf{I} + \mathbf{t}_s^e + \phi_1 \phi_s p_* \mathbf{I}, \quad p_* \equiv \rho_1 \frac{\partial A_1}{\partial \phi_1} \quad (6)$$

In the above result,  $\mathbf{I}$  is the identity,  $\mathbf{t}_s^e$  denotes the classical effective stress tensor in the sense of Terzaghi and physico-chemical effects are incorporated through the *hydration pressure* component  $p_*$ . The effective stress tensor  $\mathbf{t}_s^e$  measures stresses induced by mineral to mineral contact and primarily controls the deformation of non-swelling systems. The modified effective stress principle (6) for swelling media has the additional term,  $p_*$ . Clearly, this additional component is of physico-chemical nature and can be viewed as a stress structural component arising from the clay surface hydration forces.

*Linearized effective stress constitutive relation:*

$$\mathbf{t}_s^e = (\lambda_s \text{div} \mathbf{u}_s - \alpha_s K_s \theta) \mathbf{I} + 2\mu_s \nabla^s \mathbf{u}_s \quad (7)$$

where  $\{\mu_s, \lambda_s\}$  are the pair of Lamé coefficients of the platelet matrix,  $K_s \equiv 3\lambda_s + 2\mu_s$  denotes the solid matrix bulk modulus,  $\alpha_s$  is the coefficient of thermal expansion of the solid phase, and  $\theta \equiv T - \bar{T}$ .

*Linearized hydration stress constitutive relation*

$$\phi_1 \phi_s p_* = \bar{p}_* \bar{\phi}_1 \bar{\phi}_s + \bar{f}(\phi_1 - \bar{\phi}_1) + \frac{Q' \bar{\phi}_s}{\bar{\rho}_1} (\rho_1 - \bar{\rho}_1) - \alpha_{1s} \gamma \bar{\phi}_s \theta \quad (8)$$

where  $Q'$  is the usual poroelastic coefficient in the sense of Biot<sup>13</sup> or Biot and Wills<sup>14</sup>  $\alpha_{1s}$  is a physico-chemical coefficient of thermal expansion<sup>15</sup>  $\gamma \equiv \bar{\phi}_1 (\partial p_* / \partial \phi_1)$  denotes the bulk modulus for hydration stresses and  $\bar{f} \equiv \bar{\phi}_s \gamma + \bar{p}_* (\bar{\phi}_s - \bar{\phi}_1)$ .

*Modified Darcy's law for the adsorbed water:*

$$q_1 = -\bar{K}_1(\nabla p_1 + \bar{p}_* \nabla \phi_1) \quad (9)$$

where  $\bar{K}_1$  denotes the hydraulic conductivity of the clay particles. This shows that, in addition to a pressure gradient, the hydrophilic attraction between vicinal water and solid leads to an additional driving force (volume fraction gradient) for vicinal water flow.

*Energy equation for the mixture:*

$$\text{div} \mathbf{h} - \bar{\rho} \hat{H} + \bar{\rho} C \frac{\partial \theta}{\partial t} = -\bar{T}(\alpha_{1s} \gamma \bar{\phi}_s + K_s \alpha_s) \text{div} \frac{\partial \mathbf{u}_s}{\partial t} + \frac{\bar{T} \alpha_1 \bar{\phi}_1}{\bar{\rho}_1 \beta_1} \frac{\partial \rho_1}{\partial t} \quad (10)$$

where  $\mathbf{h}$  is the overall heat flux,  $\hat{H}$  denotes an external heat supply,  $\bar{\rho}$  denotes overall particle density at the initial equilibrium state,  $C$  is the constant volume specific heat and  $\beta_1$  and  $\alpha_1$  are the compressibility and coefficient of thermal expansion of the adsorbed water. The terms in the r.h.s. of (10) measure respectively the mechanical work associated with hydration stresses, effective stresses and fluid compressibility.

*Fourier's law of heat conduction:*

$$\mathbf{h} = -\bar{K}_T \nabla \theta \quad (11)$$

where  $\bar{K}_T$  is the particle thermal conductivity.

*Linearized equation of state of the adsorbed water:*

$$p_1 = \bar{p}_1 + \frac{Q'}{\bar{\phi}_1} (\phi_1 - \bar{\phi}_1) + \frac{1}{\bar{\rho}_1 \beta_1} (\rho_1 - \bar{\rho}_1) + \frac{\alpha_1}{\beta_1} \theta \quad (12)$$

The above system consists of the set of two-scale governing equations in terms of the unknowns  $\{\phi_1, \mathbf{u}_s, \rho_1, \mathbf{q}_1, \mathbf{t}, p, p_1, p_s, \mathbf{t}_s^e, p_*, \theta, \mathbf{h}\}$ . Further, if we multiply the mass balances of the solid phase (2) by  $\bar{\rho}_1$  and the total mass conservations (3) by  $\bar{\phi}_s$ , and add them up we obtain the mass balance of the adsorbed fluid (in terms of the Darcy velocity  $\mathbf{q}_1$ ).

$$\bar{\phi}_1 \bar{\phi}_s \frac{\partial \rho_1}{\partial t} + \bar{\rho}_1 \frac{\partial \phi_1}{\partial t} + \bar{\rho}_1 \bar{\phi}_s \text{div} \mathbf{q}_1 = 0 \quad (13)$$

If we eliminate  $\{\mathbf{t}, \mathbf{t}_s^e, \mathbf{q}_1, \mathbf{h}, p, p_s\}$  using the constitutive equations (6), (7), (9), (11), (4) and (5) then the above system can be rewritten as

$$\mu_s \Delta \mathbf{u}_s + (\lambda_s + \mu_s) \nabla \text{div} \mathbf{u}_s - \nabla p_1 + \nabla(\phi_1 \phi_s p_*) - \alpha_s K_s \nabla \theta + \bar{\phi}_s \bar{\mu}_* \nabla \frac{\partial \phi_1}{\partial t} = 0$$

$$\frac{\bar{\phi}_1}{\bar{\rho}_1} \frac{\partial \rho_1}{\partial t} + \text{div} \frac{\partial \mathbf{u}_s}{\partial t} - \bar{K}_1 (\Delta p_1 + \bar{p}_* \Delta \phi_1) = 0$$

$$\bar{K}_T \Delta \theta + \bar{\rho} \hat{H} = \bar{\rho} C \frac{\partial \theta}{\partial t} + \bar{T}(\alpha_{1s} \gamma \bar{\phi}_s + K_s \alpha_s) \text{div} \frac{\partial \mathbf{u}_s}{\partial t} - \frac{\bar{T} \alpha_1 \bar{\phi}_1}{\bar{\rho}_1 \beta_1} \frac{\partial \rho_1}{\partial t}$$

with  $\phi_1$  as in (2). Together with the constitutive relations (8) and (12) this formulates the problem in terms of the unknowns  $\{\rho_1, \mathbf{u}_s, \phi_1, p_*, p_1, \theta\}$ .

Assume now that induced changes in  $\rho_1$  due to  $p_1$  and  $p_*$  are small compared to those induced by changes in  $\theta$  (this weak dependence of  $\rho_1$  on  $p_*$  due to physico-chemical interaction was experimentally verified by Low,<sup>16</sup> who observed that changes in  $\rho_1$  due to the proximity of the solid are negligible compared to the other properties). Under these assumptions set  $\beta_1 \rightarrow 0$  and  $Q' \rightarrow 0$  and thus (12) and (8) reduce to

$$\rho_1 - \bar{\rho}_1 = -\bar{\rho}_1 \alpha_1 \theta \quad (14)$$

$$\phi_1 \phi_s p_* = \bar{\phi}_1 \bar{\phi}_s \bar{p}_* + \bar{f}(\phi_1 - \bar{\phi}_1) - \alpha_{1s} \gamma \bar{\phi}_s \theta \quad (15)$$

Using the mass balance of the solid phase  $\phi_1 - \bar{\phi}_1 = \bar{\phi}_s \operatorname{div} \mathbf{u}_s$  one can express the above governing equations in terms of  $\{\mathbf{u}_s, p_1, \theta\}$  as

$$\mu_s \Delta \mathbf{u}_s + (\lambda_s + \mu_s + \bar{\phi}_s \bar{f}) \nabla \operatorname{div} \mathbf{u}_s - \nabla p_1 + \bar{\phi}_s^2 \bar{\mu}_* \nabla \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t} - (\alpha_s K_s - \alpha_{1s} \gamma \bar{\phi}_s) \nabla \theta = 0 \quad (16)$$

$$-\bar{\phi}_1 \alpha_1 \frac{\partial \theta}{\partial t} + \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t} - \bar{K}_1 (\Delta p_1 + \bar{\phi}_s \bar{p}_* \Delta \operatorname{div} \mathbf{u}_s) = 0 \quad (17)$$

$$\bar{K}_T \Delta \theta + \bar{\rho} \hat{H} = \left( \bar{\rho} C + \frac{\bar{T} \alpha_1^2 \bar{\phi}_1}{\beta_1} \right) \frac{\partial \theta}{\partial t} + \bar{T} (\alpha_{1s} \gamma \bar{\phi}_s + K_s \alpha_s) \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t} \quad (18)$$

An important consequence of the above system is the reproduction of the classical linear model of thermoelastic consolidation (see e.g. Reference 9) upon neglect of the physico-chemical coefficients  $\{\bar{p}_*, \bar{\mu}_*, \alpha_{1s} \bar{f}\}$ .

### 2.1. Coupled particle–bulk water mesoscopic system

Let  $\Omega_f$  be the bulk phase domain occupied by a slightly compressible bulk water. In the absence of convective effects, the linearized mass balance, Darcy's law, equation of state  $p_f = p_f(\theta_f, \rho_f)$  and Fourier's law are

$$\frac{\partial \rho_f}{\partial t} + \bar{\rho}_f \operatorname{div} \mathbf{v}_f = 0 \quad (19)$$

$$\mathbf{v}_f = -\bar{K}_f \nabla p_f \quad \text{in } \Omega_f \quad (20)$$

$$p_f = \frac{1}{\bar{\rho}_f \beta_f} (\rho_f - \bar{\rho}_f) + \frac{\alpha_f}{\beta_f} \theta_f \quad (21)$$

$$\mathbf{h}_f = -\bar{K}_{Tf} \nabla \theta_f \quad (22)$$

where the corresponding bulk water properties are denoted by the subscript f. The above system together with the energy equation completes the description of the bulk-water-governing equations.

The coupled two-scale mesoscopic model can be obtained by supplementing the clay particle and bulk water systems by initial conditions and boundary conditions on the particles–bulk phase interface  $\Gamma$ . Continuity of mass, temperature and normal component of the overall stress

tensor on the boundary leads to (neglecting convective effects)

$$\mathbf{q}_l \cdot \mathbf{n} = \left( \mathbf{v}_f - \frac{\partial \mathbf{u}_s}{\partial t} \right) \cdot \mathbf{n} \quad \text{on } \Gamma \quad (23)$$

$$\mathbf{t}\mathbf{n} = \mathbf{t}_f \mathbf{n} \quad \text{on } \Gamma \quad (24)$$

$$\theta = \theta_f \quad \text{on } \Gamma \quad (25)$$

when  $\mathbf{n}$  is the unit normal exterior to  $\Omega_f$ . Moreover, following References 17, 18 an additional boundary condition for the hydrodynamical problem is postulated based on continuity of the chemical potential of the fluid phase  $\mu_\alpha \equiv A_\alpha + \rho_\alpha^{-1} p_\alpha$  ( $\alpha = l, f$ ) whose gradient is the driving force for fluid flow, i.e. at equilibrium the chemical potential is constant (see References 1, 12, 19). Hence postulate

$$\mu_l = \mu_f \quad \text{on } \Gamma \quad (26)$$

The above boundary condition can be used to evaluate the discontinuity in the liquid pressure on  $\Gamma$ . To quantify this recall our constitutive assumptions  $A_l = A_l(\theta, \rho_l, \phi_l)$  and define the entropy of the vicinal water  $\eta_l \equiv -\partial A_l / \partial \theta$ . Using the chain rule, this together with the chemical potential definition and definitions (4) and (6) for  $p_l$  and  $p_*$  yields

$$d\mu_l = d\left(\frac{p_l}{\rho_l}\right) + dA_l = d\left(\frac{p_l}{\rho_l}\right) + \frac{p_l}{\rho_l^2} d\rho_l - \eta_l d\theta + \frac{p_*}{\rho_l} d\phi_l = \frac{1}{\rho_l} (d\rho_l + p_* d\phi_l) - \eta_l d\theta \quad (27)$$

which is the Gibbs–Duhem relation for the vicinal liquid. This, together with the boundary conditions (25) and (26) imply  $d\mu_l = d\theta = 0$  on  $\Gamma$  and thus

$$dp_l = -p_* d\phi_l \quad \text{on } \Gamma \quad (28)$$

Denote  $\phi_l^*$  the value of the volume fraction for which hydration and swelling forces are absent, (see References 12, 20, 21). For  $\phi_l = \phi_l^*$  the intra-particle water behaves as a bulk fluid and thus we have  $p_l(\phi_l^*) = p_f$ . Therefore, integrating (28) we get

$$p_l - p_f = \int_{\phi_l}^{\phi_l^*} p_*(s) ds \quad \text{on } \Gamma \quad (29)$$

In contrast to non-swelling media, the above result shows a discontinuity in the fluid pressure across the interface as a consequence of the hydration pressure  $p_*$ . For non-swelling media ( $p_* = 0$ ) we have  $p_l = p_f$  which shows continuity in fluid pressure on the interface between a non-swelling medium and a free fluid.

For small strains, the linearized form of (29) about the initial equilibrium state is

$$p_l - p_f = \int_{\phi_l}^{\phi_l^*} \bar{p}_*(s) ds - \bar{p}_*(\phi_l - \bar{\phi}_l) \quad \text{on } \Gamma \quad (30)$$

### 3. THREE-SCALE MASS TRANSFER EQUATION BETWEEN ADSORBED AND BULK WATER

In this section we couple the two-scale model in the particle domain  $\Omega_l$  with the equations for bulk water flow in  $\Omega_f$  and proceed within and additional upscaling towards the derivation of

some three-scale results. This is accomplished within the homogenization procedure. In particular, we conduct our approach towards a rigorous derivation of a generalized mass transfer equation between adsorbed and bulk water, similarly to the one postulated by Ma and Hueckel.<sup>6-8</sup>

### 3.1. Homogenization

Within the framework of homogenization,<sup>5</sup> our macroscopic swelling clay is idealized as a bounded domain  $\Omega^\varepsilon$  with a periodic structure. We then introduce mesoscopic and macroscopic lengths, denoted by  $l$  and  $L$ , which characterize the mesoscopic size of the period and the macroscopic field, respectively. Their ratio  $\varepsilon \equiv l/L$ . Consider  $\Omega^\varepsilon$  as the union of disjoint parallelepiped cells,  $Q^\varepsilon$ , congruent to a standard  $Q$  consisting of the union of several clay particles  $Q_1$  completely surrounded by a connected bulk water domain  $Q_f$ . Let the systems of bulk phase water and clay particles in  $\Omega^\varepsilon$  be denoted by  $\Omega_f^\varepsilon$  and  $\Omega_1^\varepsilon$ , respectively. The  $\varepsilon$ -model in  $\Omega^\varepsilon$  consists of the aforementioned two-scale particle and bulk-phase-governing equations in each subdomain  $\Omega_1^\varepsilon$  and  $\Omega_f^\varepsilon$ . Our starting point,  $\varepsilon = 1$  corresponds to our unscaled mesoscopic model where (16)–(18) and (19)–(22) hold. For  $\varepsilon < 1$ , a swelling clay soil is posited wherein the centres of the bulk-phase channels are located  $\varepsilon$ -times the reference distance apart, though congruent to the reference cell (Figure 1). The homogenized model governing the macroscopic clay soil is obtained by letting  $\varepsilon \rightarrow 0$  while the lattice extends to infinity. The picture corresponding to the limiting model is depicted in Figure 2, where a mesoscopic cell  $Q$  is assigned to each point  $\mathbf{x}$  of the macroscopic bulk-phase domain. This leads to a macroscopic model of *dual porosity* type wherein the swelling clay soil is macroscopically represented as two porous distinct structures coexisting at each macroscopic point: one representing the clay particles and the other representing the bulk water. In this picture the macroscopic Darcy velocity is that of the bulk water alone and the fluid storage occurs in the system of clay particles. This technique has been successfully used to model naturally fractured reservoirs in which the system of fractures play the role of the bulk-phase system (where the macroscopic flow takes place) and the matrix blocks behave as the analogue of the clay particles (see Reference 22). The main feature of dual porosity approaches is the accurate description of the interaction (mass transfer) between the two structures. Within the framework of homogenization this mass transfer (also referred to as secondary flux) arises naturally as a source term in the mass balance of the fracture flow appearing distributed over the whole macroscopic domain. (see References 23, 24, 22).

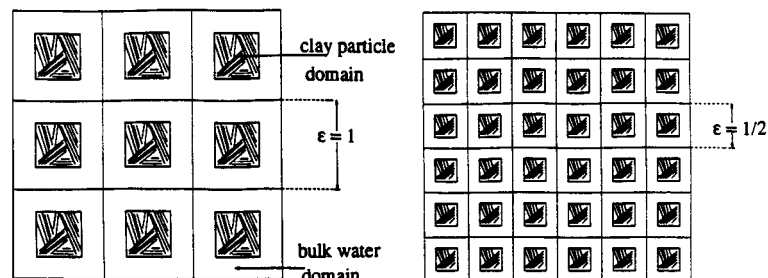


Figure 1. Two elements of equivalent clay soils



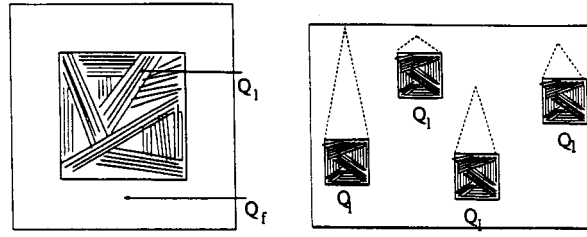


Figure 2. The reference cell  $Q$  and its distribution over the homogenized soil

We shall consider a totally fissured medium in the sense of Douglas and Arbogast<sup>22</sup> wherein particles are completely isolated from each other by the bulk phase fluid. In this picture there is no direct mass and momentum transfer from particle to particle. Instead the adsorbed water first flows into the bulk phase where it passes into another particle or remains in the bulk phase.

A crucial point in the derivation of dual porosity models by homogenization is the proper scaling of the permeability of the clay particles  $\bar{K}_1$  by an appropriate power of  $\varepsilon$ . The idea is to conserve flow in some sense and consequently avoid degeneration of the governing equations as  $\varepsilon \rightarrow 0$ . Following Arbogast and co-workers<sup>22,23</sup> for naturally fractured reservoirs or Murad and Cushman<sup>18</sup> for swelling clays, this is done by considering the scaling law  $\bar{K}_1^f = \bar{K}_1 \varepsilon^2$ . This scaling has the effect of making the particles progressively less permeable as  $\varepsilon \rightarrow 0$  and consequently preserves the mass transfer function.

The formal process is accomplished by considering every property to be of the form  $f(\mathbf{x}, \mathbf{y})$  (where  $\mathbf{x}$  and  $\mathbf{y}$  denote the macroscopic and mesoscopic coordinates respectively (with  $\mathbf{y} = \varepsilon^{-1} \mathbf{x}$ ) and then postulating two scale asymptotic expansions for the set  $\mathbf{u}^\varepsilon$  consisting of the unknowns  $\{\mathbf{u}_s, \mathbf{q}_1, \phi_1, p_1, \rho_1, \mathbf{t}, \mathbf{h}, \theta, \mathbf{t}_s^e, p_*\}$  and  $\{\rho_f, \mathbf{v}_f, p_f, \theta_f, \mathbf{h}_f\}$  in terms of the perturbation parameter  $\varepsilon$

$$\mathbf{u}^\varepsilon = \mathbf{u}^0 + \varepsilon \mathbf{u}^1 + \varepsilon^2 \mathbf{u}^2 + \dots$$

with the coefficients  $\mathbf{u}^i$ ,  $\Omega$ -periodic in  $\mathbf{y}$ . Inserting the above developments into the set of governing mesoscopic equations and noting that by the chain rule the differential operator  $\partial/\partial_{\mathbf{x}}$  is replaced by  $\partial/\partial_{\mathbf{x}} + \varepsilon^{-1} \partial/\partial_{\mathbf{y}}$  we obtain the following results:

For the overall momentum equation (16), denoting  $\lambda_s^* = \lambda_s + \mu_s + \bar{\phi}_s \bar{f}$

$$\begin{aligned} & [\mu_s (\text{div}_{\mathbf{x}} + \varepsilon^{-1} \text{div}_{\mathbf{y}}) (\nabla_{\mathbf{x}} + \varepsilon^{-1} \nabla_{\mathbf{y}}) + \lambda_s^* (\nabla_{\mathbf{x}} + \varepsilon^{-1} \nabla_{\mathbf{y}}) (\text{div}_{\mathbf{x}} + \varepsilon^{-1} \text{div}_{\mathbf{y}})] [\mathbf{u}_s^0 + \varepsilon \mathbf{u}_s^1 + \dots] \\ & + [\bar{\mu}_* \bar{\phi}_s (\nabla_{\mathbf{x}} + \varepsilon^{-1} \nabla_{\mathbf{y}}) (\text{div}_{\mathbf{x}} + \varepsilon^{-1} \text{div}_{\mathbf{y}})] \left[ \frac{\partial \mathbf{u}_s^0}{\partial t} + \varepsilon \frac{\partial \mathbf{u}_s^1}{\partial t} + \dots \right] \\ & - (\nabla_{\mathbf{x}} + \varepsilon^{-1} \nabla_{\mathbf{y}}) (p_1^0 + \varepsilon p_1^1 + \dots) \\ & - [(\alpha_s K_s + \alpha_{l,s} \gamma \bar{\phi}_s) (\nabla_{\mathbf{x}} + \varepsilon^{-1} \nabla_{\mathbf{y}})] [\theta^0 + \varepsilon \theta^1 + \dots] = 0 \end{aligned}$$

For Darcy's law (9)

$$(\mathbf{q}_1^0 + \varepsilon \mathbf{q}_1^1 + \dots) = -\varepsilon^2 \bar{K}_1 [\nabla_{\mathbf{x}} + \varepsilon^{-1} \nabla_{\mathbf{y}}] [p_1^0 + \varepsilon p_1^1 + \dots + \bar{p}_* (\phi_1^0 + \varepsilon \phi_1^1 + \dots)]$$

For the mass balances (3) and (2)

$$\begin{aligned} \frac{\bar{\phi}_1}{\bar{\rho}_1} \frac{\partial}{\partial t} (p_1^0 + \varepsilon p_1^1 + \dots) + (\operatorname{div}_x + \varepsilon^{-1} \operatorname{div}_y) \left( \frac{\partial \mathbf{u}_s^0}{\partial t} + \varepsilon \frac{\partial \mathbf{u}_s^1}{\partial t} + \dots + \mathbf{q}_1^0 + \varepsilon \mathbf{q}_1^1 + \dots \right) &= 0 \\ \frac{1}{\bar{\phi}_s} \frac{\partial}{\partial t} (\phi_1^0 + \varepsilon \phi_1^1 + \dots) - (\operatorname{div}_x + \varepsilon^{-1} \operatorname{div}_y) \left( \frac{\partial \mathbf{u}_s^0}{\partial t} + \varepsilon \frac{\partial \mathbf{u}_s^1}{\partial t} + \dots \right) &= 0 \end{aligned}$$

For the effective stress relation (7)

$$\begin{aligned} \mathbf{t}_s^0 + \varepsilon \mathbf{t}_s^1 + \dots &= [\lambda_s (\operatorname{div}_x + \varepsilon^{-1} \operatorname{div}_y) \mathbf{I} + 2\mu_s (\nabla_x^s + \varepsilon^{-1} \nabla_y^s)] (\mathbf{u}_s^0 + \varepsilon \mathbf{u}_s^1 + \dots) \\ &\quad - \alpha_s K_s (\theta^0 + \varepsilon \theta^1 + \dots) \mathbf{I} \end{aligned} \quad (31)$$

By a formal matching of the powers of  $\varepsilon$  we obtain successive cell problems.

$$O(\varepsilon^{-2}) \quad \mu_s \Delta_{yy} \mathbf{u}_s^0 + \lambda_s^* \nabla_y \operatorname{div}_y \mathbf{u}_s^0 = 0 \quad (32)$$

$$O(\varepsilon^0) \quad \mathbf{q}_1^0 = 0$$

$$O(\varepsilon^1) \quad \mathbf{q}_1^1 = -\bar{K}_1 (\nabla_y p_1^0 + \bar{p}_* \nabla_y \phi_1^0)$$

$$O(\varepsilon^0) \quad \frac{\bar{\phi}_1}{\bar{\rho}_1} \frac{\partial \rho_1^0}{\partial t} + \operatorname{div}_y \mathbf{q}_1^1 + \operatorname{div}_x \frac{\partial \mathbf{u}_s^0}{\partial t} + \operatorname{div}_y \frac{\partial \mathbf{u}_s^1}{\partial t} = 0 \quad (33)$$

$$O(\varepsilon^0) \quad \frac{1}{\bar{\phi}_s} \frac{\partial \phi_1^0}{\partial t} - \operatorname{div}_x \frac{\partial \mathbf{u}_s^0}{\partial t} - \operatorname{div}_y \frac{\partial \mathbf{u}_s^1}{\partial t} = 0 \quad (34)$$

Applying the same procedure to the bulk water gives

$$\begin{aligned} \frac{1}{\bar{\rho}_f} \frac{\partial}{\partial t} (\rho_f^0 + \varepsilon \rho_f^1 + \dots) + (\operatorname{div}_x + \varepsilon^{-1} \operatorname{div}_y) (\mathbf{v}_f^0 + \varepsilon \mathbf{v}_f^1 + \dots) &= 0 \\ \mathbf{v}_f^0 + \varepsilon \mathbf{v}_f^1 + \dots &= -\bar{K}_f (\nabla_x + \varepsilon^{-1} \nabla_y) (p_f^0 + \varepsilon p_f^1 + \dots) \\ p_f^0 + \varepsilon p_f^1 + \dots + \frac{1}{\bar{\rho}_f \beta_f} ((\rho_f^0 + \varepsilon \rho_f^1 + \dots) - \bar{\rho}_f) + \frac{\alpha_f}{\beta_f} (\theta_f^0 + \varepsilon \theta_f^1 + \dots) \\ \mathbf{h}_f^0 + \varepsilon \mathbf{h}_f^1 + \dots &= -\bar{K}_{\text{Tr}} (\nabla_x + \varepsilon^{-1} \nabla_y) (\theta_f^0 + \varepsilon \theta_f^1 + \dots) \end{aligned}$$

After collecting powers of  $\varepsilon$  this yields

$$O(\varepsilon^0) \quad \frac{1}{\bar{\rho}_f} \frac{\partial \rho_f^0}{\partial t} + \operatorname{div}_x \mathbf{v}_f^0 + \operatorname{div}_y \mathbf{v}_f^1 = 0 \quad (35)$$

$$O(\varepsilon^0) \quad \rho_f^0 = \frac{1}{\bar{\rho}_f \beta_f} (\rho_f^0 - \bar{\rho}_f) + \frac{\alpha_f}{\beta_f} \theta_f^0 \quad (36)$$

$$O(\varepsilon^{-1}) \quad \nabla_y p_f^0 = \nabla_y \theta_f^0 = 0 \quad (37)$$

Using the same procedure to the boundary conditions (23) gives

$$O(\varepsilon^1) \quad \mathbf{q}_l^1 \cdot \mathbf{n} = \left( \mathbf{v}_f^1 - \frac{\partial \mathbf{u}_s^1}{\partial t} \right) \cdot \mathbf{n} \quad \text{on } \Gamma \quad (38)$$

In addition using (31) in the boundary condition (24) and noting that  $\mathbf{t}_f = -p_f \mathbf{I}$  we get

$$O(\varepsilon^{-1}) \quad (2\mu_s \nabla_y^s \mathbf{u}_s^0 + \lambda_s \operatorname{div}_y \mathbf{u}_s^0 \mathbf{I}) \mathbf{n} = 0 \quad \text{on } \Gamma \quad (39)$$

From (37) we have  $p_f^0 = p_f^0(\mathbf{x}, t)$  and  $\theta_f^0 = \theta_f^0(\mathbf{x}, t)$ . Together with (36) this yields  $\rho_f^0 = \rho_f^0(\mathbf{x}, t)$ . Also note that  $\mathbf{u}_s^0$  satisfies the Neumann problem given by (32) and (39), whose solution is  $\mathbf{u}_s^0 = \mathbf{u}_s^0(\mathbf{x}, t)$ , which shows that the leading order neighbouring platelets move together as a rigid body on each cell.

We now derive the overall macroscopic mass balance. Begin by introducing the local mean value operator

$$\widetilde{\cdot} = |Q|^{-1} \int_{Q_i} \cdot dQ_i, \quad i = 1, f$$

By averaging (35) and (33) using the boundary conditions (38) along with the divergence theorem and the periodicity assumption we obtain (recall that  $\mathbf{n}$  was chosen outward to  $Q_1$ )

$$\begin{aligned} \frac{n_f}{\bar{\rho}_f} \frac{\partial \rho_f^0}{\partial t} + \widetilde{\operatorname{div}_x \mathbf{v}_f^0} &= -|Q|^{-1} \int_{Q_f} \operatorname{div}_y \mathbf{v}_f^1 dQ_f = |Q|^{-1} \int_{\Gamma} \mathbf{v}_f^1 \cdot \mathbf{n} d\Gamma \\ &= |Q|^{-1} \int_{\Gamma} \left( \mathbf{q}_l^1 + \frac{\partial \mathbf{u}_s^1}{\partial t} \right) \cdot \mathbf{n} d\Gamma \\ &= |Q|^{-1} \int_{Q_1} \left( \operatorname{div}_y \mathbf{q}_l^1 + \operatorname{div}_y \frac{\partial \mathbf{u}_s^1}{\partial t} \right) dQ_1 = -(1 - n_f) \operatorname{div}_x \frac{\partial \mathbf{u}_s^0}{\partial t} - \frac{\bar{\phi}_1}{\bar{\rho}_1} \frac{\partial \bar{\rho}_1^0}{\partial t} \end{aligned}$$

where  $n_\alpha = |Q_\alpha|/|Q|$ ,  $\alpha = 1, f$  denotes the macroscopic volume fractions of the particles and bulk phase. Hence, defining the averaged Darcy velocity of the bulk phase relative to the solid phase by  $\tilde{\mathbf{q}}_f^0 \equiv \tilde{\mathbf{v}}_f^0 - n_f \partial \mathbf{u}_s^0 / \partial t$  we have

$$n_f \frac{\partial \rho_f^0}{\partial t} + \bar{\rho}_f \left( \frac{\bar{\phi}_1}{\bar{\rho}_1} \frac{\partial \bar{\rho}_1^0}{\partial t} + \operatorname{div}_x \frac{\partial \mathbf{u}_s^0}{\partial t} + \operatorname{div}_x \tilde{\mathbf{q}}_f^0 \right) = 0 \quad (40)$$

The above result is our total macroscopic mass balance. Next, we combine this result with the macroscopic mass balance for the bulk phase water in order to derive our main three-scale result: *the generalized mass transfer equation between adsorbed and bulk water.*

### 3.2. Mass Transfer Equation

The macroscopic bulk water balance can be derived using the averaging theorem which relates the spatial average of a time derivative to the derivative of a spatial average. This theorem was used by Withaker<sup>25</sup> who denominated it a local Reynolds Transport Theorem (see also References 26 and 27). Let thus  $f^\varepsilon(\mathbf{x}, t)$  be a general scalar mesoscopic function defined on  $Q_1^\varepsilon$  and let

$\partial \mathbf{u}_s^\varepsilon / \partial t$  denote the mesoscopic velocity of particle bulk phase interface  $\Gamma^\varepsilon$  (neglecting convective effects). From Reference 25 the local Reynolds Transport Theorem can be expressed as

$$\frac{1}{|Q^\varepsilon|} \frac{\partial}{\partial t} \int_{Q_i^\varepsilon} f^\varepsilon dQ_i^\varepsilon = \frac{1}{|Q^\varepsilon|} \int_{Q_i^\varepsilon} \frac{\partial f^\varepsilon}{\partial t} dQ_i^\varepsilon + \frac{1}{|Q^\varepsilon|} \int_{\Gamma^\varepsilon} f^\varepsilon \frac{\partial \mathbf{u}_s^\varepsilon}{\partial t} \cdot \mathbf{n} d\Gamma \quad (41)$$

Note that the above result is scale-dependent since the domain of integration  $Q_i^\varepsilon$  depends on  $\varepsilon$ . Thus, a proper scaling law of the terms is mandatory. To derive the correct scaling law we make use of a change of variables and compute the integral over the reference unit cell  $Q_1$ . For given  $\varepsilon$ , denote the three-dimensional and two-dimensional Jacobian  $J_Q^\varepsilon$  and  $J_\Gamma^\varepsilon$  of the transformations that map  $Q_1$  onto  $Q_i^\varepsilon$  and  $\Gamma$  onto  $\Gamma^\varepsilon$ , respectively. It is well known that  $J_Q^\varepsilon$  and  $J_\Gamma^\varepsilon$  govern, respectively, changes in volume ( $J_Q^\varepsilon = |Q_i^\varepsilon|/|Q_1| = \varepsilon^3$ ) and area ( $J_\Gamma^\varepsilon = |\Gamma^\varepsilon|/|\Gamma| = \varepsilon^2$ ). We then have

$$\begin{aligned} \int_{Q_i^\varepsilon} f^\varepsilon dQ_i^\varepsilon &= \int_{Q_1} f^\varepsilon J_Q^\varepsilon dQ_1 = \varepsilon^3 \int_{Q_1} f^\varepsilon dQ_1 \\ \int_{\Gamma^\varepsilon} f^\varepsilon d\Gamma^\varepsilon &= \int_\Gamma f^\varepsilon J_\Gamma^\varepsilon d\Gamma = \varepsilon^2 \int_\Gamma f^\varepsilon J_\Gamma^\varepsilon d\Gamma \end{aligned}$$

Using the above transformations one can evaluate (41) in the reference domain  $Q_1$

$$\frac{1}{|Q|} \frac{\partial}{\partial t} \int_{Q_1} f^\varepsilon dQ_1 - \frac{1}{|Q|} \int_{Q_1} \frac{\partial f^\varepsilon}{\partial t} dQ_1 = \frac{1}{\varepsilon|Q|} \int_\Gamma f^\varepsilon \frac{\partial \mathbf{u}_s^\varepsilon}{\partial t} \cdot \mathbf{n} d\Gamma \quad (42)$$

Consider now the expansions for  $f^\varepsilon(\mathbf{x})$  and  $\mathbf{u}_s^\varepsilon(\mathbf{x})$

$$f^\varepsilon = f^0(\mathbf{x}, \mathbf{y}, t) + \varepsilon f^1(\mathbf{x}, \mathbf{y}, t) + \dots$$

$$\mathbf{u}_s^\varepsilon = \mathbf{u}_s^0(\mathbf{x}, t) + \varepsilon \mathbf{u}_s^1(\mathbf{x}, \mathbf{y}, t) + \dots$$

where we have used the fact that  $\mathbf{u}_s^0 = \mathbf{u}_s^0(\mathbf{x}, t)$ . Since the above result holds for arbitrary  $f^\varepsilon$  set  $f^\varepsilon$  independent of  $\mathbf{y}$  and  $t$ . Thus, choosing  $f^\varepsilon(\mathbf{x}, \mathbf{y}, t) = f^0(\mathbf{x})$  we get for the two terms on l.h.s. of (42)

$$\begin{aligned} \frac{1}{|Q|} \int_{Q_1} \frac{\partial f^\varepsilon}{\partial t} dQ_1 &= \frac{1}{|Q|} \int_{Q_1} \frac{\partial f^0(\mathbf{x})}{\partial t} dQ_1 = 0 \\ \frac{1}{|Q|} \frac{\partial}{\partial t} \int_{Q_1} f^\varepsilon dQ_1 &= f_0(\mathbf{x}) \frac{\partial n_1}{\partial t} \end{aligned}$$

Using the above results, (42) reduces to

$$f_0(\mathbf{x}) \frac{\partial n_1}{\partial t} = \frac{f_0(\mathbf{x})}{\varepsilon|Q|} \int_\Gamma \left( \frac{\partial \mathbf{u}_s^0}{\partial t}(\mathbf{x}, t) + \varepsilon \frac{\partial \mathbf{u}_s^1}{\partial t} + \dots \right) \cdot \mathbf{n} d\Gamma$$

By collecting the powers  $\varepsilon^{-1}$  and  $\varepsilon^0$  along with the divergence theorem we get

$$\frac{1}{|Q|} \int_{\Gamma} \frac{\partial \mathbf{u}_s^0}{\partial t}(\mathbf{x}, t) \cdot \mathbf{n} d\Gamma = 0$$

$$\frac{\partial n_1}{\partial t} = \frac{1}{|Q|} \int_{\Gamma} \frac{\partial \mathbf{u}_s^1}{\partial t} \cdot \mathbf{n} d\Gamma = \frac{1}{|Q|} \int_{Q_1} \operatorname{div}_y \frac{\partial \mathbf{u}_s^1}{\partial t} dQ_1$$

Hence, since  $n_f = 1 - n_1$ , using (34) we obtain

$$\frac{\partial n_f}{\partial t} = -\frac{1}{|Q|} \int_{Q_1} \operatorname{div}_y \frac{\partial \mathbf{u}_s^1}{\partial t} dQ_1 = n_1 \operatorname{div}_x \frac{\partial \mathbf{u}_s^0}{\partial t} - \frac{1}{\bar{\phi}_s} \frac{\partial \tilde{\phi}_1^0}{\partial t}$$

where  $\widetilde{\operatorname{div}_x \mathbf{u}_s^0}(\mathbf{x}, t) = n_1 \operatorname{div}_x \mathbf{u}_s^0$ . In conjunction with (40) this yields

$$n_1 n_f \frac{\partial \rho_f^0}{\partial t} + \bar{\rho}_f \frac{\partial n_f}{\partial t} + \bar{\rho}_f n_1 \widetilde{\operatorname{div}_x \mathbf{q}_f^0} = -\frac{\bar{\rho}_f n_1 \bar{\phi}_1}{\bar{\rho}_1} \frac{\partial \tilde{\rho}_1^0}{\partial t} - \frac{\bar{\rho}_f}{\bar{\phi}_s} \frac{\partial \tilde{\phi}_1^0}{\partial t} \quad (43)$$

The above is our *three-scale fluid mass balance*. A notable consequence of this result is its resemblance with the two-scale fluid mass balance (13) except the r.h.s. where (43) exhibits an additional source term which accounts for the interchange mass transfer between adsorbed and bulk water. This mass transfer source term exhibits two different components: one due to changes in adsorbed water density and another due to changes in the intra-particle porosity. The latter component is somewhat related to the mass transfer of Ma and Hueckel.<sup>6</sup> In our formulation this effect is measured by the changes in the averaged volume fraction (the last term in the r.h.s. of (43)). According to Ma and Hueckel<sup>8</sup> the first term in the r.h.s. (involving  $\partial \tilde{\rho}_1^0 / \partial t$ ) is much less significant than the mass transfer term (involving  $\partial \tilde{\phi}_1^0 / \partial t$ ) and therefore can be neglected. A remarkable consequence of the homogenization procedure for derivation of the three-scale dual porosity model is that it provided a rational basis for the appearance of the mass transfer term. Therefore there is no need to postulate *a priori* the existence of this quantity. On the other hand, its evaluation is more complex as it requires the solution of the intra-particle two-scale problem (16)–(18) in the particle domain  $\Omega_1$ .

The microstructural approach underlying the generalized inter-phase mass transfer concept implies in a strong dependence of the r.h.s. of (43) on clay geometry. Highly compacted clays, whose structure appears dominated by narrow spaces,<sup>28</sup> are characterized by the large size of the domain of periodic cell (clay particles) relative to the void domain (1–5  $\mu\text{m}$ ). Hence, the cell-averaging in the r.h.s. of (43) is performed over a larger domain and consequently has a strong influence on the macroscopic bulk-phase flow. Due to the narrow void spacing the bulk phase water flows at very low percolation rates<sup>29</sup> and consequently the macroscopic hydrodynamics of very dense smectite clays is mainly dictated by the adsorbed water. On the other hand, the size of the periodic cell in loose clays, is relatively small compared to the wide void spacing ((2–50  $\mu\text{m}$ )) and thus high percolation rates related to the bulk phase flow dominates the macroscopic water transport through the system. In the derivation of the three-scale results within the homogenization procedure, the cluster structure appears explicitly through the geometry of the periodic cell where the averaging is performed. Different cluster structures lead to different cell geometries and consequently to different macroscopic behaviors.

A simplified constitutive equation for the mass transfer resembling in form the one postulated by Ma and Hueckel, can be obtained if we make use of a time-scale assumption in the particle domain and consider that intra-particle transient effects related to the net hydration effective stress component  $p_* + \text{tr } \mathbf{t}_s^e$  are neglected. The idea underlying this time-scale assumption is to consider that net hydration-effective stress component reaches equilibrium instantaneously as it is disturbed by the bulk phase, i.e.  $\partial(p_* + \text{tr } \mathbf{t}_s^e)/\partial t \rightarrow 0$ . We proceed by illustrating that this assumption leads to a linear dependence between the time rates of change of volume fraction and temperature and thus reproduces in form the mass transfer equation postulated by Ma and Hueckel. To show this take the trace in (7), use (2) and add to (15) to obtain

$$\frac{\partial}{\partial t} (\text{tr } \mathbf{t}_s^e + \phi_1 \phi_s p_*) = \left( \frac{K_s}{\bar{\phi}_s} + \bar{f} \right) \frac{\partial \phi_1}{\partial t} - (3\alpha_s K_s + \alpha_{1,s} \gamma \bar{\phi}_s) \frac{\partial \theta}{\partial t}$$

Making use of the aforementioned time-scale assumption the above l.h.s. vanishes and hence we have after averaging

$$O(\varepsilon^0) \quad \left( \frac{K_s}{\bar{\phi}_s} + \bar{f} \right) \frac{\partial \bar{\phi}_1^0}{\partial t} = (3\alpha_s K_s + \alpha_{1,s} \gamma \bar{\phi}_s) \frac{\partial \bar{\theta}^0}{\partial t}$$

in which after using in the last term of the r.h.s. of (43) leads to a mass transfer equation, depending linearly on temperature rate, therefore resembling in form the one postulated by Ma and Hueckel.

#### 4. VARIATIONAL FORMULATION AND FINITE ELEMENT APPROXIMATION

We now discretize the two-scale governing equations (16)–(18) and illustrate their application to a highly compacted clay buffers with a monomodal distribution composed of clay minerals and adsorbed water. To this end consider an alternative formulation of the mesoscopic problem. Following References 1, 12 this formulation consists in introducing a new auxiliary variable defined in the particle domain referred to as *local reference bulk-phase pressure*  $p_B$ . To characterize locally this auxiliary variable consider a fixed solid strain and define  $p_B$  as the thermodynamic pressure of a virtual bulk fluid at instantaneous thermal and mechanical equilibrium with the adsorbed water (same chemical potentials and temperature) whose properties are unaffected by the solid phase (i.e. no physico-chemical interaction between the reference bulk fluid and the clay surface). Let the properties of this virtual reference bulk fluid in  $\Omega_i$  be denoted by the subscript  $B$  to distinguish from the subscript  $f$  used for the bulk fluid in the bulk phase domain  $\Omega_f$ . Since by construction  $\mu_B \equiv \mu_1$  and  $\theta_B \equiv \theta$ , by the same arguments based on integration of the Gibbs–Duhem relation (27) leading to (29) we get

$$p_1 - p_B = \int_{\phi_1}^{\phi_1^*} p_*(s) ds \quad (44)$$

Hence for given  $\phi_1$ ,  $p_1$  and  $\phi_1^*$  (44) can be used to define locally the reference bulk-phase pressure  $p_B$ . Together with the solid-phase mass balance (2), the linearized version of (44) is

$$p_1 - p_B = \int_{\phi_1}^{\phi_1^*} \bar{p}_*(s) ds - \bar{p}_*(\phi_1 - \bar{\phi}_1) = \int_{\phi_1}^{\phi_1^*} \bar{p}_*(s) ds - \bar{p}_* \bar{\phi}_s \text{div } \mathbf{u}_s \quad (45)$$

Note that the term on the r.h.s. of (44) allow us to quantify the perturbation in the vicinal water pressure  $p_1$  relative to  $p_B$  due to physico-chemical interaction with the clay surface. This fact was explored in Murad<sup>1</sup> who introduced the following quantities: (1) The excess in adsorbed water pressure relative to the reference bulk-phase pressure,  $\delta_p \equiv p_1 - p_B$ . (2) The excess in pore pressure relative to the reference bulk phase  $\Pi_B \equiv p - p_B$ . (3) The swelling pressure  $\Pi \equiv -(\mathbf{t} - \mathbf{t}_B)$ , where  $\mathbf{t}_B \equiv -p_B \mathbf{I} + \mathbf{t}_s^c$  is the total particle stress tensor defined in the sense of the classical Terzaghi's effective stress principle (in the absence of physico-chemical forces, where particles are viewed as a mixture of clay platelets and reference bulk fluid). All the above definitions reflect locally the excess in the vicinal water properties due to the physico-chemical interaction with the minerals. In other words the above quantities would be zero if the properties of the water are not perturbed by the presence of the platelets, ( $p_* = 0$ ). Note that using the definitions for  $\mathbf{t}_B$  and  $\Pi_B$  and equation (6) for  $\mathbf{t}$  in the swelling pressure definition leads to  $\Pi = p - p_B - \phi_1 \phi_s p_* = \Pi_B - \phi_1 \phi_s p_*$ . This shows that  $\Pi$  is a scalar and the hydration pressure can be viewed as the difference between  $\Pi_B$  and  $\Pi$ . This in conjunction with (6) yields

$$\mathbf{t} = -(p - p_* \phi_1 \phi_s) \mathbf{I} + \mathbf{t}_s^c = -(p_B + \Pi_B - p_* \phi_1 \phi_s) \mathbf{I} + \mathbf{t}_s^c = -(p_B + \Pi) \mathbf{I} + \mathbf{t}_s^c \quad (46)$$

Equation (46) consists of an alternative form of writing the mesoscopic modified effective stress principle (6) with  $p$  replaced by  $p_B$ . In this case physico-chemical forces appeared measured by the swelling pressure  $\Pi$ .

For simplicity assume that the vicinal water is incompressible ( $\alpha_1 \rightarrow 0$ ). Thus, using (45), in terms of  $\{\mathbf{u}_s, p_B, \theta\}$  our two-scale governing equations (16)–(18) can be rewritten as

$$\mu_s \Delta \mathbf{u}_s + (\lambda_s^* + \mu_s) \nabla \operatorname{div} \mathbf{u}_s - \nabla p_B + \mu^* \nabla \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t} - \alpha^* \nabla \theta = 0 \quad (47)$$

$$\operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t} - \bar{K}_1 \Delta p_B = 0 \quad (48)$$

$$\bar{K}_T \Delta \theta + \bar{\rho} \hat{H} = \bar{\rho} C \frac{\partial \theta}{\partial t} + \bar{T} \alpha^* \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t} \quad (49)$$

where now  $\lambda_s^* \equiv \lambda_s + \bar{\phi}_s (\bar{f} + \bar{p}_*)$ ,  $\mu^* = \bar{\phi}_s^2 \bar{\mu}_*$  and  $\alpha^* = \alpha_s K_s + \alpha_{ls} \bar{\phi}_s \gamma$ . In addition, using (44), boundary condition (29) can be rewritten as

$$p_B = p_t \quad \text{on } \Gamma \quad (50)$$

Note that in contrast to (29) the formulation (47)–(49) in terms of  $\{\mathbf{u}_s, p_B, \theta\}$  implies in continuity in the reference bulk fluid pressure on the particle–bulk phase interface. Note that within this formulation, after solving the above system in terms of  $\{\mathbf{u}_s, p_B, \theta\}$  we can easily compute  $p_1$  using the post processing (45).

We now solve numerically the system (47)–(49) aiming applications to a argillaceous repository containing a heat-emitting radioactive waste. For this particular application consider that the surrounding bulk fluid is at atmospheric pressure ( $p_t = 0$ ) and for simplicity also consider homogeneous interface conditions of Dirichlet type on the boundary of the clay buffer and initial conditions of the type  $\theta = \hat{\theta}$  and  $\operatorname{div} \mathbf{u} = 0$ . This latter initial condition expresses the incompressible response of the solid–fluid aggregate in the beginning of the consolidation process. This

imposes an internal constraint to the problem close to  $t = 0$  and consequently leads to a variational formulation consisting in finding a weak solution of a saddle-point problem whose approximation gives rise to a mixed finite element method.

#### 4.1. Weak form

For simplicity, consider two-dimensional problems. Let  $L^2(\Omega)$  denote the usual set of square integrable scalar valued functions defined on  $\Omega_1$ , with inner product

$$(f, g) \equiv \int_{\Omega} fg \, d\Omega \quad \forall f, g \in L^2(\Omega)$$

and let the corresponding space of vector functions  $\mathbf{L}^2(\Omega) \equiv (L^2(\Omega))^2$ . Further let  $H_0^1(\Omega)$  be the space of scalar functions

$$H_0^1(\Omega) \equiv \{f \in L^2(\Omega), \nabla f \in \mathbf{L}^2(\Omega), f = 0 \text{ on } \Gamma\}$$

and  $\mathbf{H}_0^1(\Omega) \equiv (H_0^1(\Omega))^2$ . The variational formulation for (47)–(49) is given by

For each  $t \in (0, \infty)$ , find  $\{\mathbf{u}_s(t), p_B(t), \theta(t)\} \in \mathbf{H}_0^1(\Omega) \times H_0^1(\Omega) \times H_0^1(\Omega)$  such that

$$2\mu_s(\mathbf{E}_s(\mathbf{u}_s), \mathbf{E}_s(\mathbf{v})) + \left( \lambda_s^* \operatorname{div} \mathbf{u}_s + \mu^* \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t}, \operatorname{div} \mathbf{v} \right) - (p_B - \alpha^* \theta, \operatorname{div} \mathbf{v}) = 0 \quad \forall \mathbf{v} \in \mathbf{H}_0^1(\Omega)$$

$$\left( \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t}, q \right) + \bar{K}_1(\nabla p_B, \nabla q) = 0 \quad \forall q \in H_0^1(\Omega)$$

$$\bar{K}_T(\nabla \theta, \nabla \tau) + \bar{\rho} C \left( \frac{\partial \theta}{\partial t}, \tau \right) + \alpha^* \bar{T} \left( \operatorname{div} \frac{\partial \mathbf{u}_s}{\partial t}, \tau \right) = \bar{\rho}(\hat{H}, \tau) \quad \forall \tau \in H_0^1(\Omega)$$

with  $\mathbf{E}_s(\mathbf{u}_s)$  given as in (1). For given  $\{p_B, \mathbf{u}_s\}$  weak solutions of the above problem, the adsorbed water pressure  $p_1$  can be evaluated using the weak form of the post-processing (45)

$$(p_1, q) = (p_B, q) + (\bar{\Pi}_B, q) - \bar{p}_* \bar{\phi}_s(\operatorname{div} \mathbf{u}_s, q) \quad \forall q \in L^2(\Omega)$$

### 5. FINITE ELEMENT APPROXIMATION

We now turn to the approximate solution by finite elements. For simplicity admit that  $\Omega_1$  is a polygonal domain discretized by a uniform mesh of  $N_e$  non-overlapping elements. Let thus  $U_h^k$  and  $V_h^l$  be families of continuous finite-dimensional subspaces of  $H_0^1(\Omega)$ , whose interpolations functions reduced to polynomials of degree  $k$  and  $l$ , on each triangle or quadrilateral of a partition of  $\Omega_1$ . Define a completely discrete solution based on the backward Euler method in time and a Galerkin method using continuous approximating functions in space. Let  $\Delta t$  be the time step and let  $\mathbf{u}^n$  denotes the solution  $\mathbf{u}(t)$  at time  $t_n = n\Delta t$ . The backward Euler method replaces the time derivative  $\partial \mathbf{u} / \partial t$  by the backward difference quotient  $\partial_t \mathbf{u}^n = (\mathbf{u}^n - \mathbf{u}^{n-1}) / \Delta t$ . It is then natural to



consider the following backward Euler Galerkin method:

For each  $t \in (0, \infty)$ , find  $\{\mathbf{u}_h^m, p_h^m, \theta_h^m\} \in \mathbf{U}_h^k \times V_h^l \times U_h^k$  such that

$$\begin{aligned} 2\mu_s(\mathbf{E}_s(\mathbf{u}_h^m), \mathbf{E}_s(\mathbf{v}_h)) + \lambda^*(\operatorname{div} \mathbf{u}_h^m, \operatorname{div} \mathbf{v}_h) + \frac{\mu^*}{\Delta t}(\operatorname{div} \mathbf{u}_h^m - \operatorname{div} \mathbf{u}_h^{m-1}, \operatorname{div} \mathbf{v}_h) - (p_h^m, \operatorname{div} \mathbf{v}_h) \\ - \alpha^*(\theta_h^m, \operatorname{div} \mathbf{v}_h) = 0 \quad \forall \mathbf{v}_h \in \mathbf{U}_h^k \end{aligned} \quad (51)$$

$$\frac{1}{\Delta t}(\operatorname{div} \mathbf{u}_h^m - \operatorname{div} \mathbf{u}_h^{m-1}, q_h) + \bar{K}_1(\nabla p_h^m, \nabla q_h) = 0, \quad \forall q_h \in V_h^l \quad (52)$$

$$\frac{\bar{\rho}C}{\Delta t}(\theta_h^m - \theta_h^{m-1}, \tau_h) + \bar{K}_T(\nabla \theta_h^m, \nabla \tau_h) + \frac{\alpha^* \bar{T}}{\Delta t}(\operatorname{div} \mathbf{u}_h^m - \operatorname{div} \mathbf{u}_h^{m-1}, \tau_h) = \bar{\rho}(\hat{H}, \tau_h) \quad \forall \tau_h \in U_h^k \quad (53)$$

If we consider absence of relaxation effects initially and that for given  $\theta_m^0 = \hat{\theta}$ , the initial values  $\{\mathbf{u}_h^0, p_h^0\}$  satisfy (51) and the initial condition  $\operatorname{div} \mathbf{u}_h^0 = 0$ , then we are led to solve the following incompressible elasticity (for Stokes flow) problem

$$2\mu(\mathbf{E}_s(\mathbf{u}_h^0), \mathbf{E}_s(\mathbf{v}_h)) - (p_h^0, \operatorname{div} \mathbf{v}_h) = \alpha^*(\hat{\theta}, \operatorname{div} \mathbf{v}_h) \quad \forall \mathbf{v}_h \in \mathbf{U}_h^k \quad (54)$$

$$(\operatorname{div} \mathbf{u}_h^0, q_h) = 0 \quad \forall q_h \in V_h^l \quad (55)$$

It is well known that the stability of the above mixed finite element formulation is ensured provided the finite element spaces  $\mathbf{U}_h^k$  and  $V_h^l$  satisfy Babuška–Breezi (BB) or *inf-sup* condition<sup>30</sup>

$$\sup_{\mathbf{v}_h \in \mathbf{U}_h^k} \frac{(\operatorname{div} \mathbf{v}_h, q_h)}{\|\nabla \mathbf{v}_h\|} \geq C \|q_h\|, \quad \forall q_h \in V_h^l \quad (56)$$

The choice  $k = l + 1$ , corresponding to Taylor–Hood elements<sup>31</sup> (first proposed for incompressible flows) verifies (56) and leads to stable and optimally convergent solution of (54) and (55). Unstable approximations such as when displacement and pressure approximations are piecewise polynomials of the same degree ( $k = l$ ) do not fulfill the BB condition. Consequently these methods show a lack of stability, exhibiting oscillations in the pore pressure approximation for small time where nearly incompressible deformations in the porous matrix prevail.<sup>32,33</sup> The convergence analysis of this type of problem exhibits singular behavior close to  $t = 0$ .<sup>32–35</sup>

## 6. NUMERICAL RESULTS

Analytical solutions for thermoelastic systems resembling in form, our two-scale governing equations have neglected the effect of mechanical work on the energy equation and consequently solved the thermoelastic problem as two uncoupled problems (see e.g. Reference 36). In what follows, we shall numerically simulate the fully coupled problem in dimensionless form and illustrate numerically the influence of physico-chemical effects through the magnitude of the dimensionless numbers involved.

Let thus  $L$  be a characteristic length and, without loss of generality assume that Poisson's coefficient of the buffer matrix vanishes, i.e.  $\lambda_s = 0$ . Introduce the following dimensionless quantities:

$$\begin{aligned} \mathbf{x}_i^* &= \frac{\mathbf{x}_i}{L}, & \mathbf{U} &= \frac{\mathbf{u}_s}{L}, & t^* &= \frac{\mu_s \bar{K}_1 t}{L^2}, & P_B &= \frac{p_B}{\mu_s}, & \theta^* &= 2\theta\alpha_s \\ N_1 &= \frac{K_T}{2\alpha_s \mu_s^2 \bar{K}_1}, & N_2 &= \frac{\bar{\rho} C}{\mu_s \alpha_s}, & H^* &= \frac{\rho \hat{H} L^2}{\bar{K}_1 \mu_s^2} \\ PC &= \frac{\lambda_s^*}{\mu_s}, & REL &= \frac{\bar{\mu}_* \bar{K}_1}{L^2}, & PC_\theta &= \frac{\alpha^*}{\alpha_s K_s} - 1 \end{aligned}$$

Without loss of generality, assume  $N_1 = N_2 = 1$  and  $\bar{T} = 1/(2\alpha_s)$ . The effect of hydration forces shall be quantified in terms of the magnitude of the dimensionless numbers PC, REL and  $PC_\theta$ . The number  $PC_\theta$  measures the quotient between physico-chemical and mechanical thermal expansions of the buffer. The number PC governs the ratio between physico-chemical and effective mechanical effects under isothermal consolidation. Small values of PC are typical of coarse-grained soils such as sands, silts and low and non-expansive medium plastic clays such as kaolinite or illite. On the other hand, active plastic smectitic clays such as bentonite and montmorillonite, whose deformation is primarily controlled by physico-chemical activity, are governed by high values of PC. The number REL governs the magnitude of relaxation and viscoelastic behaviour of the adsorbed water. Clearly REL is directly related to the viscosity coefficient  $\mu_*$ . In other words,  $REL = \mu_* = 0$  if the viscosity of vicinal water is not altered by the proximity to the solid-surfaces. If we make use of the superscript “\*” to denote differentiation with respect to the dimensionless space variable  $\mathbf{x}_i^*$ , then in terms of the above quantities the fully discrete system (51)–(53) can be rewritten as

For each  $t^* \in (0, \infty)$ , find  $\{\mathbf{U}_h^m, P_h^m, \theta_h^{*m}\} \in \mathbf{U}_h^k \times V_h^l \times U_h^k$  such that

$$\begin{aligned} 2(\mathbf{E}_s^*(\mathbf{U}_h^m), \mathbf{E}_s^*(\mathbf{v}_h)) + \left( \frac{REL}{\Delta t^*} + PC \right) (\text{div}^* \mathbf{U}_h^m, \text{div}^* \mathbf{v}_h) - (P_h^m \text{div}^* \mathbf{v}_h) \\ - (1 + PC_\theta)(\theta_h^{*m}, \text{div}^* \mathbf{v}_h) = \frac{REL}{\Delta t^*} (\text{div}^* \mathbf{U}_h^{m-1}, q_h), \quad \forall \mathbf{v}_h \in \mathbf{U}_h^k \\ (\text{div}^* \mathbf{U}_h^m, q_h) + \Delta t^* (\nabla^* P_h^m, \nabla^* q_h) = (\text{div}^* \mathbf{U}_h^{m-1}, q_h), \quad \forall q_h \in V_h^l \\ (\theta_h^{*m}, \tau_h) + \Delta t^* (\nabla^* \theta_h^{*m}, \nabla^* \tau_h) + (1 + PC_\theta)(\text{div}^* \mathbf{U}_h^m, \tau_h) = (\theta_h^{*(m-1)}, \tau_h) \\ + (1 + PC_\theta)(\text{div}^* \mathbf{U}_h^{m-1}, \tau_h) + \Delta t^* (H^*, \tau_h), \quad \forall \tau_h \in U_h^k \end{aligned}$$

In order to perform numerical simulations of the above system we select a problem related to a clay-based engineered barrier for encapsulating a heat generating container. Depicted in Figure 3 the example consists of a heat source embedded in a bentonitic clay buffer region which is compacted in a borehole located in a granite block. The heater carries a decaying source, modelled as a distributed volumetric heat production  $\hat{H}(t)$  exponentially decreasing in time,  $\hat{H}(t) = \hat{H}_0 \exp(-\omega t)$  where  $\omega$  is the decreasing constant and  $\hat{H}_0$  is the initial power.

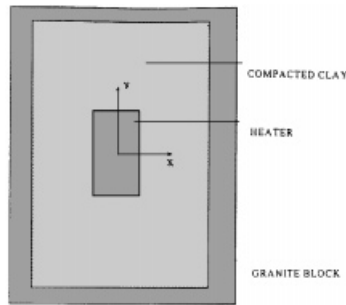


Figure 3. Decreasing heat source buried in a compacted clay

Coupled thermomechanical and hydraulic effects are thermally induced by the heater on the clay buffer. The difference between the thermal expansions of the adsorbed water and clay matrix (under coupled mechanical and physico-chemical effects) gives rise to the generation of positive excess pore pressures in the vicinity of the heat source. Since we have assumed that the vicinal fluid is incompressible in order to maintain the same effects consider the case where the overall coefficient of thermal expansion of the platelet matrix  $\alpha_s + \alpha_{ls}$  is negative which means that an increase in temperature produces compressive volumetric strains and generate positive pore pressures. This behaviour has been explained due to irreversible structural adjustments of the platelet matrix (see Reference 15 for details). Hence, the localized excess in liquid pressure produces a water flow through the clay buffer which is directed away from the zone of heating. The fluid movement is followed by a consolidation process in the clay and consequently by a liquid pressure dissipation. Next, we illustrate numerically the effect of hydration forces on the performance of the barrier through the magnitude of the numbers PC and REL. For simplicity, we concentrate our simulations in the case where  $PC_0 = 0$ .

Assume that the highly compacted buffer exhibits monomodal adsorbed water distribution. Consider that granite–buffer interface behaves as a perfect semipermeable membrane for the hydraulic problem and in addition, assume that the stiffness of the granite block is much larger than that of the buffer such that, for the purpose of our simulations, the granite may be considered rigid. Assume that the time scale of the thermomechanical process in the granite is much smaller than that of the buffer such that the granite is at thermal and hydraulic equilibrium with its surroundings (assumed at zero pressure and temperature). The aforementioned time scale and rigidity assumptions on the granite block allow us to postulate  $\theta = P_B = 0$  on the granite–buffer interface. The purpose of these simplifications is to concentrate our numerical results on the buffer and consequently better illustrate the influence of physico-chemical effects. Due to the symmetry we consider a fourth domain of the buffer (Figure 3) which is discretized by a finite element mesh of rectangular elements. To fulfil the stability condition dictated by the inf-sup condition close to  $t = 0$ , we adopt the lowest order of Taylor–Hood family, consisting of  $15 \times 15$  biquadratic elements for displacements and temperature and bilinear for pore pressure. The subsequent figures depicts the influence of physico-chemical effects in terms of the difference between total pressure  $P = -(\frac{1}{3}) \text{tr} \mathbf{t}$  and bulk-phase pressure  $p_B$ . This excess is denoted by  $E$ . Taking the trace in (46) we have

$$E \equiv -\frac{1}{3} \text{tr} \mathbf{t} - p_B = P - p_B = P_s^c + \Pi$$

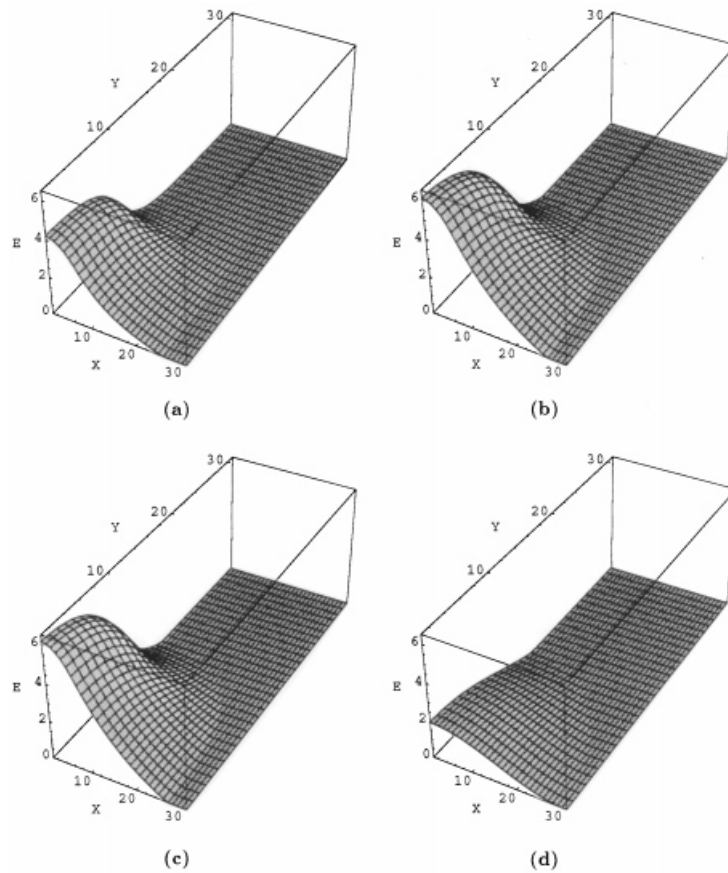


Figure 4. Influence of physico-chemical effects in terms of the excess in total pressure relative to bulk phase pressure:  $E = P - P_B = P_s^e + \Pi$ : (a)  $PC = REL = 0$ ; (b)  $PC = REL = 10$ ; (c)  $PC = REL = 50$ ; (d) Swelling pressure elevation  $\Pi = E - P_s^e$

where  $P_s^e \equiv -(\frac{1}{3})\text{tr} \mathbf{t}_s^e$  is the effective pressure. Figure 4 (a)–(c) depicts the spatial distribution of the excess  $E$  for three values of  $PC = REL = 0, 10, 50$ , respectively, and  $t$  fixed. For  $PC = REL = 0$  we have  $\Pi = 0$  and  $E = P_s^e$  which shows that in the absence of physico-chemical effects the excess  $E$  is nothing but the effective pressure. For positive  $PC$  and  $REL$ , the quantity  $E$  incorporates both effective and swelling pressure contributions. Figure 4(d) depicts the difference  $E - P_s^e = \Pi$  for  $PC = REL = 50$ . We may observe that the physico-chemical influence due to  $\Pi$  is larger close to the heat source. Figure 5 illustrates the comparison between the spatial distributions of  $p_1$  and  $p_B$  relative to the initial excess in pore pressure  $\bar{\Pi}_B$ , i.e. after solving the discrete problem for  $p_B$  and  $\mathbf{u}_s$  we compute  $p_1 - \bar{\Pi}_B = p_B - p_* \phi_s \text{div} \mathbf{u}_s$  in the discrete version of the post-processing (45). The first case (Figure 5(a)) exhibits no physico-chemical effects,  $\bar{p}_* = \bar{\Pi}_B = 0$  and  $p_1 = p_B$ . The second plot (Figure 5(b)) exhibits physico-chemical effects ( $\phi_s \bar{p}_* = 2$ ). Same behaviour observed in Figure 4 is also valid, i.e. physico-chemical effects amplify the magnitude of  $p_1$  especially close to the heater. Finally we illustrate the influence of relaxation

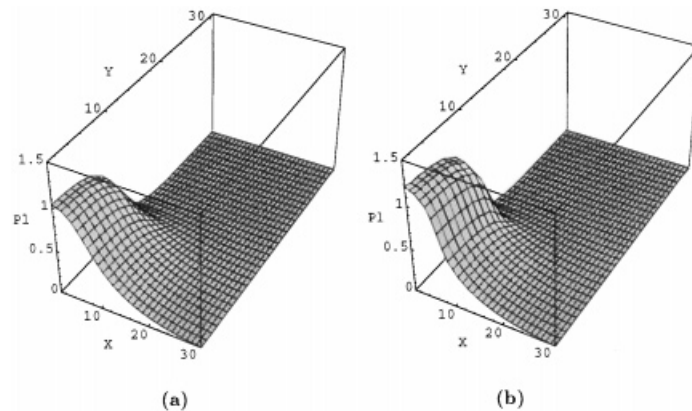


Figure 5. Liquid pressure elevation: (a) Absence of physico-chemical effects  $\bar{p}_* = 0$ ,  $p_1 = p_B$ ; (b) With physico-chemical effects,  $\bar{p}_* > 0$ ,  $p_1 > p_B$

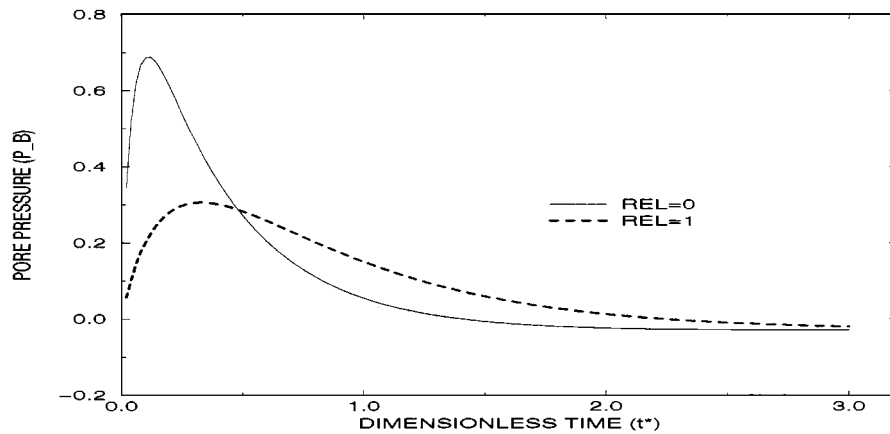


Figure 6. Evolution in time of the adsorbed liquid pressure

effects on the evolution of the pore pressure. Figure 6 depicts the evolution of  $p_B$ , averaged over the region occupied by the heater, as a function of the dimensionless time  $t^*$ , for two values of REL. In the first plot neither relaxation nor physico-chemical effects are present ( $PC = REL = 0$ ). The dashed line illustrates the same evolution for  $REL = PC = 1$ . Relaxation effects smooth out the sharp layer in the pressure for small time. We also note the effects of REL on the long-term behaviour are manifest in the slower asymptotic decay compared to the case where  $REL = 0$ .

## CONCLUSIONS

The two-scale HMT model of Murad<sup>1</sup> for highly compacted clays composed of clay minerals and adsorbed water was solved numerically by finite elements. Variational principles and the Galerkin approximation is space combined with the backward Euler time-stepping method in time

were proposed to discretize the two-scale HMT governing equations. Numerical simulations of a argillaceous repository containing a heat-emitting radioactive waste were performed showing the influence of physico-chemical forces on the performance of the clay buffer.

Partial results toward a three-scale thermomechanical macroscopic model including the bulk phase next to the swelling particles were derived by homogenizing the two-scale model with the bulk water. A notable consequence of the upscaling process is that it provided a rational basis for the appearance of a generalized mass transfer function between adsorbed and bulk water. The ideas underlying this mass transfer we introduced by Ma and Hueckel<sup>6</sup> who postulated its existence as a source in fluid mass balance. The three-scale homogenized model consists of a first attempt at a rigorous derivation of a mass transfer equation which incorporates intra-particle mobility of the adsorbed water.

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